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KINETICS AND MODELING OF OIL EXTRACTION FROM VIETNAM LEMONGRASS BY STEAM DISTILLATION

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ABSTRACT

Essential oils from parts of plants such as stump, flower, kernel, and seed are usually produced by extraction, distillation and mechanical press. In practice, steam distillation is commonly used for the extraction of crude essential oils since it is not only a simple process but also applicable at various scales. Furthermore, the method makes it possible for a keeping of precious components of the oils unchanged. Therefore, studies on kinetics and modeling of the essential oil steam distillation are needed for the optimization of the operating conditions, energy requirement, and the process scale-up.

In this work, experiments of lemongrass (*Cymbopogon Citratus*) steam distillation were carried out and a kinetics model was developed for the extraction of lemongrass essential oil. Raw materials were pretreated by natural drying, primarily crushing and chopping prior to the distillation. The oil yield obtained is in the range of 2.1 – 2.9 ml/kg of raw materials. Composition of the crude oil extracted was measured by GC-MS. Measurements showing that the oil product contains 70.5 % of precious component–Citral in which Neral is 29.8 % and Geranial 40.7 %. The kinetics parameters were estimated from experimental data conducted at various operating conditions for different preparation of the raw materials. The process rate constant (extraction rate constant) describing the extraction efficiency obtained from this study is varied from 0.02 to 0.027 min^{-1} using first-order kinetic model.

Keywords: Lemongrass, essential oil, steam distillation, kinetics, modeling.

1. INTRODUCTION

Lemongrass (*Cymbopogon Citratus*) is a perennial tropical grass, which is commonly cultivated for essential oil in several countries such as India, Brazil, Mexico, and Vietnam. Cultivation of this plant can be carried out at different geographic regions due to its high resistant to climatic variation and plagues. The essential oil extracted from lemongrass is often used in pharmaceutical and cosmetic fields [1].

According to literature, essential oils can be extracted by both traditional and novel techniques such as solvent extraction, steam distillation, hydrodistillation, and supper critical CO_2 extraction. Each method has its particular advantages and disadvantages [2, 3]. Comparison between the two methodologies is still debated because the proper extraction process is dependent on various parameters and criteria. Among the aforementioned methods, steam distillation is commonly used for essential oil extraction due to safety, simple and environmental-friendly operations. In addition, high quality oil can be obtained without decomposition since the oil vapors are entrained by the steam flows during the extraction process and the surrounding water is a barrier to keep the materials off overheating or charring [4].

There have been numerous studies dealing with the yield, composition and biological activities of essential oils obtained by steam distillation from different plant species grown all over the world. However, the kinetics of essential oil steam distillation has been studied to a much smaller extent despite its importance not only for the fundamental understanding but also for operation, optimization, control and design of industrial steam distillation processes. Kinetic models along with essential oil yield and composition are important for water-steam distillation processes from both technological and economical viewpoints. Therefore, this work focuses on a study of kinetic modeling of Vietnam lemongrass water-steam distillation.

2. EXPERIMENTS

2.1. Plant materials

Fresh lemongrass was procured from a local supplier in Thanh Oai, Hanoi. The raw materials (stems) were 10 cm in length. In order to prepare for the extraction, raw materials were further treated for certain experiments. The pre-treatment of the virgin materials was aimed to examine the effects of feedstock size and initial water content on the oil yield. Five experiments carried out at different characteristics of the treated materials were shown in Table 1.

Table 1. Characteristics of treated lemongrass.

	Material Characteristics
Exp. 1	2 cm, fresh, chopped grass
Exp. 2	2 cm, natural dry*, chopped grass
Exp. 3	2 cm, Crushed, fresh, chopped grass
Exp. 4	2 cm, crushed, cut, fresh, chopped grass
Exp. 5	2 cm, crushed, cut, natural dry, chopped grass

*: Raw material was dried naturally in shadow for 48 hours

2.2. Experimental setup

The experimental lab-scale system for water-steam distillation of lemongrass is shown in Figure 1. The designed capacity of the still stripper is 15 kg per batch. The mixture of treated lemongrass and water is heated using a 6 kW electrical resistant heater and the system is operated at atmospheric pressure.

The equipment was fitted with a tight lid to prevent oil and vapor from leaking out. The system is operated in a manner that steam rising from the still stripped the oil away from the plant material and the vapor comprising oil and steam was passed to a condenser where the vapor phase was condensed and separated. In the decanter (the oil – water separator), essential oil was separated from water at the top of the separator since the density of the oil is lighter than that of water.

Figure 1. Steam distillation unit: (1) Electrical heater; (2) Hydrostatic level; (3) Distillation still; (4, 5): Vapor lines; (6) Condenser; (7) Oil – water separator.

For each experiment, 8.5 – 9.5 kg of treated lemongrass was loaded into the still. The distillation was carried out for total 180 minutes in which the oil recovery was measured at proper extraction time for kinetic modeling study. Maximum accumulated oil obtained from the five experiments was also recorded for the analysis of oil recovery. In this work, the oil yield is calculated as the ratio between the total oil obtained and mass of the raw material loaded (ml/kg) [5].

2.4. GC-MS conditions

Composition of the extracted essential oil from lemongrass was analyzed by gas chromatography–mass spectrometry (GC-MS). The analysis of the essential oils was performed in the capillary column (30 m, 0.32 mm i.d., 0.25 film thickness). Column temperature was initially 40 ^{o}C for 2 minutes, and then gradually increased to 225 ^{o}C at the rate of 4 ^{o}C/ min. The extracts were diluted 3:100 (v/v) with acetone 99.99 %. Temperature of the injector and detector were set at 290 ^{o}C and 175 ^{o}C, respectively. Split ratio was set at 1:100 and the carrier gas was helium operated at a flow rate of 2.2 ml/min.

3. KINETIC MODELING

In this section, the kinetics of essential oil extraction from lemongrass was developed based on experimental data and the first-order kinetic model. According to literature [4, 6, 7], several kinetic models of essential oil extraction from plants have been reported. However, a general kinetic modeling of the essential oils by steam- and/or hydro-distillations considering both tissue diffusion and surface transportation of the oils is still under development. For design and scale-up purpose, this work focuses on the apparent kinetic model of Vietnam lemongrass steam distillation in which the rate-determining step is considered as the transportation of the oils from feedstock surfaces into the vapor phase. Therefore, the rate at which oil is extracted from lemongrass is directly proportional to the amount of oil present in the plant material at any time. The relationship can be expressed mathematically in the form of a first order ordinary differential equation (ODE).

$$\frac{dC}{dt} = -kC \qquad (1)$$

where: C – average quantity of essential oil in the grass at any time t, ml; k – first order rate constant, min^{-1}; t – time of steam distillation, min.

This ODE can be solved using the variable separable method and Equation (1) can be rearranged in the following expression:

$$\frac{dC}{C} = -kdt \qquad (2)$$

In the integration form where the initial quantity of the oil is taken into account, the oil extracted ratio is expressed as a function of time and apparent extraction rate constant (k) as follows:

$$\ln\left(\frac{C}{C_0}\right) = -kt \qquad (3)$$

where C_0 – initial quantity of essential oil which was approximated to be a function of the cumulative volume of oil extracted from the lemongrass.

In order to analyze the rate of oil recovery, a new variable $y(t)$ representing the time dependent fractional extraction yield was defined as:

$$y(t) = \frac{C_0 - C}{C_0} \qquad (4)$$

From Equations (3 and 4) the fractional extraction yield is expressed as a function of time:

$$\ln\left[\frac{1}{1 - y(t)}\right] = kt \qquad (5)$$

Equation (5) is the solution of the ODE presented in Equation (1) and this is used for the determination of extraction rate constant from the experimental data.

4. RESULTS AND DISCUSSION

4.1. Effects of material characteristics

The effects of pretreated materials on the oil recovery from experimentation are addressed and discussed. The accumulated recovery of the essential oil obtained from five experiments is shown in Table 2. In experiments one and two (Exp. 1 and Exp. 2) the oil yield of 1.68 ml/kg is obtained for both fresh-chopped and natural dried-chopped materials. The oil recovery in these cases is less than that in three later experiments (see Table 2) where the raw grass was crushed in the pretreatment process. This would be explained that the oil is mainly located in the tissues of plants and the oil is hardly extracted without breaking the plant's tissues. In addition, water and steam cannot entrain the oil out of unbroken tissues during the distillation. However, in the later experiments (Exp. 3 to Exp.5) the oil yield is increased compared to that in the former ones. Therefore, the material water content does not affect the oil recovery for uncrushed feedstock while both natural drying and crushing show positive effects on the total amount of the extracts. In this work, the oil yield extracted from Vietnam lemongrass is up to 2.88 ml/kg and this range is similar to that of India lemongrass [6, 7].

Table 2. Accumulated yield of lemongrass essential oil at various material characteristics.

	Plant Sample Mass (kg)	Extraction Time (min)	Material Characteristics	Essential Oil Volume (ml)	Oil Yield (ml/kg)
Exp. 1	9.0	180	2 cm, fresh, chopped grass	15.08	1.68
Exp. 2	9.0	180	2 cm, natural dry, chopped grass	15.08	1.68
Exp. 3	9.5	180	2 cm, crushed, fresh, chopped grass	20.73	2.18
Exp. 4	9.0	180	2 cm, crushed, cut, fresh, chopped grass	20.73	2.30
Exp. 5	8.5	180	2 cm, crushed, cut, natural dry, chopped grass	24.50	2.88

4.2. Composition of Vietnam lemongrass essential oil

The essential oil obtained from lemongrass water-steam distillation in this work is analyzed by GS-MS method to determine the oil's composition. Results obtained from GC-MS analysis are depicted Figure 2 and detailed composition of the oil's components is given in Table 3.

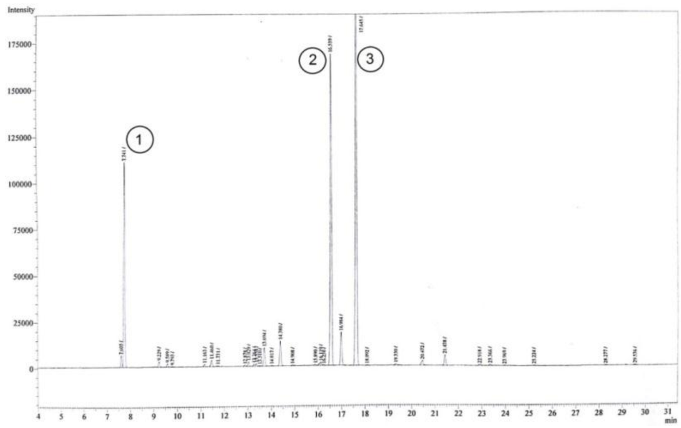

Figure 2. GC-MS analysis of lemongrass essential oil ((1): Myrcene; (2): Neral: (3): Geranial).

According to the analysis, 18 components are found in lemongrass essential oil in which Myrcene, Neral and Geranial are major ones. Detailed measurements show that the oil product contains 70.5 % of precious component–Citral in which Neral is 29.8 % and Geranial 40.7 % (see Table 3). Compared to Indian lemongrass, Citral composition in Vietnam lemongrass (70.5 %) is a little bit lower while concentration of Myrcene in Vietnam lemongrass is much higher than that in Indian lemongrass [5, 6]. This may be due to the differences in ground and climate of the two regions.

Table 3. GC/MS of lemongrass essential oil obtained by steam distillation.

No	Compound	Relative peak area (%)	Retention time (min)
1	**Myrcene**	**15,44**	**7,741**
2	Limonene	0,46	9,229
3	E,E-Cosmene	0,29	9,589
4	Z-β-Ocimene	0,22	11,163
5	E-β-Ocimene	0,67	11,460
6	α-Terpinolene	0,39	13,029
7	Citronellal	0,19	13,264
8	Cis-Verbenol	0,22	13,358
9	Linalool	1,69	13,694
10	Cis-Carveol	2,32	14,380
11	Atrimesol	0,1	14,908
12	Nerol	0,47	16,119
13	**Neral**	**29,81**	**16,559**
14	Geraniol	3,14	16,984
15	**Geranial**	**40,7**	**17,645**
16	Carveol	0,23	19,330
17	Geranyl acetate	0,62	20,472
18	Caryophellene	1,04	21,438

4.3. Kinetics study

In order to develop the first-order kinetics of the lemongrass steam distillation, the accumulated oil recovery is measured at each period of time during a batch of extraction. The oil obtained as a function of time for five experiments is indicated in Figure 3. It is seen that the oil recovery is increased with time up to about 160 minutes of distillation then it tends to reach a maximum value for each run. In this work, experimental data from experiments 3, 4, and 5 are selected for the kinetics development.

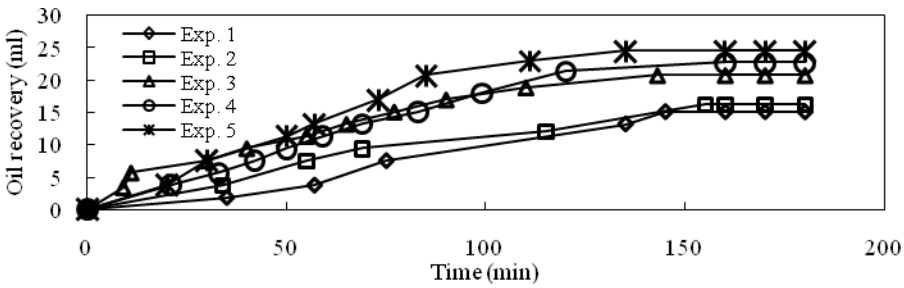

Figure 3. Oil yield versus extraction time.

Experimental data in the three experiments are interpreted in the form of time dependent fractional extraction yield as mentioned in Equation (5). It is seen that, the left hand side of the equation varies linearly with respect to extraction time. Therefore, the apparent extraction rate constant (k) is a slope of a straight line. After rearrangement of the measured data, the kinetic parameter of the lemongrass water-steam distillation is determined using polynomial fitting method. Detailed numerical results are shown in Figure 4 in comparison with measured data. The apparent extraction rate constants obtained from experiments 3, 4, and 5 are 0.02, 0.025, and 0.027 min^{-1}, respectively. These values can be used for the design and scale-up purposes.

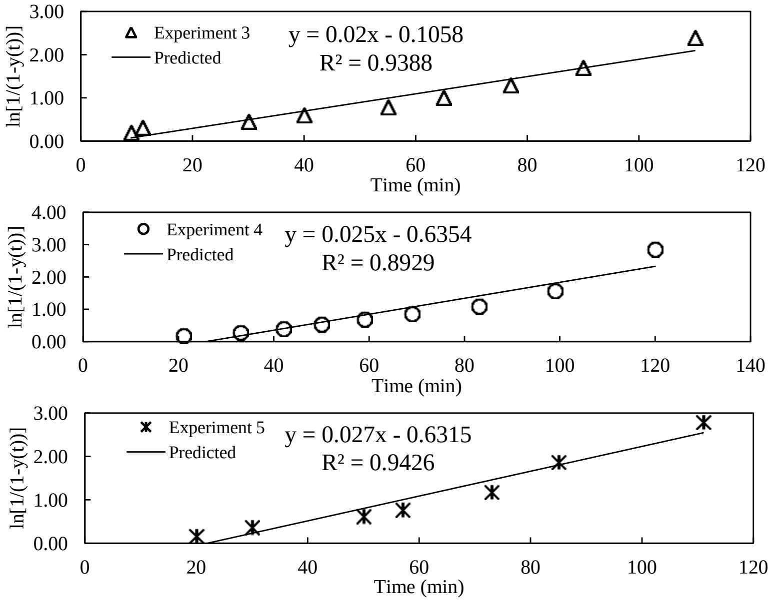

Figure 4. First-order kinetic model of lemongrass essential oil extraction.

5. CONCLUSIONS

The extraction of essential oils from Vietnam lemongrass by water-steam distillation is studied by experimentation and kinetic modeling. Expermental data show that, the oil recovery obtained from the extraction is in the range of 2.1 - 2.9 ml/kg in which crushing and natural drying pretreatments of the raw materials are positively effected on the oil recovery efficiency. GC-MS analysis shows that the oil product contains 70.5 % of precious component–citral in which neral is 29.8 % and geranial 40.7 %. The kinetics parameters are estimated from experimental data conducted at various operating conditions for different pretreatments of the raw materials. The process rate constant (extraction rate constant) describing the extraction efficiency obtained from this study is varied from 0.02 to 0.027 min^{-1} using first-order kinetic model. Based on the proposed kinetics, modeling of the essential oil water-steam distillation can be used for further application such as design and scale-up.

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TÓM TẮT

NGHIÊN CỨU QUÁ TRÌNH CHIẾT TINH DẦU SẢ CHANH BẰNG PHƯƠNG PHÁP CHƯNG LÔI CUỐN HƠI NƯỚC: THỰC NGHIỆM VÀ ĐỘNG HỌC

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Tinh dầu được tách ra từ các phần của thực vật như thân, rễ, lá, hoa và hạt bằng các phương pháp chiết, chưng cất, hoặc ép. Trong thực tế công nghiệp, chưng lôi cuốn hơi nước được sử dụng phổ biến để thu hồi tinh dầu thô do phương pháp này đơn giản, dễ triển khai với những quy mô khác nhau từ lớn đến nhỏ. Hơn nữa, phương pháp cũng có khả năng thu hồi những cấu tử quý trong tinh dầu. Chính vì vậy, nghiên cứu động học và mô hình hóa quá trình thu hồi tinh dầu bằng phương pháp chưng lôi cuốn hơi nước là rất quan trọng để tối ưu hóa quá trình công nghệ, năng lượng sử dụng, hiệu suất thu hồi tinh dầu và tính toán thiết kế chuyển quy mô sang công nghiệp. Trong nghiên cứu này, đã tiến hành thực nghiệm, xây dựng động học quá trình thu hồi tinh dầu sả chanh bằng phương pháp chưng lôi cuốn hơi nước. Kết quả thực nghiệm cho thấy lượng tinh dầu sả chanh thu được trong khoảng 2,1 – 2,9 ml/kg vật liệu thô. Thành phần của tinh dầu thu được được xác định bằng phương pháp GC–MS. Kết quả cho thấy hàm lượng Citral chiếm 70,5 % (Neral chiếm 29,8 % và Geranial chiếm 40,7 %). Các thông số động học được xác định từ các số liệu thực nghiệm với các điều kiện công nghệ khác nhau nằm trong khoảng 0,02 đến 0,027 phút^{-1}.

Từ khóa: sả chanh, tinh dầu, chưng lôi cuốn hơi nước, chiết, động học, mô hình hóa.